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EXAMINER	
LISH, PETER J	

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1754	

13

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Please find below and/or attached an Office communication concerning this application or proceeding.

9

Office Action Summary	Application No.	Applicant(s)
	09/881,684	ZHOU, OTTO Z.
	Examiner	Art Unit
	Peter J Lish	1754

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133).
- Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on 2-6-03.

2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.

3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) 1-71 is/are pending in the application.

4a) Of the above claim(s) 31,42 and 65-70 is/are withdrawn from consideration.

5)☐ Claim(s) _____ is/are allowed.

6)☒ Claim(s) 1-30, 32, 34-41, 43-64,and 71 is/are rejected.

7)☒ Claim(s) 33 is/are objected to.

8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.

10)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.

Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).

11)☐ The proposed drawing correction filed on _____ is: a)☐ approved b)☐ disapproved by the Examiner.

If approved, corrected drawings are required in reply to this Office action.

12)☐ The oath or declaration is objected to by the Examiner.

Priority under 35 U.S.C. §§ 119 and 120

13)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).

a)☐ All b)☐ Some * c)☐ None of:

1.☐ Certified copies of the priority documents have been received.

2.☐ Certified copies of the priority documents have been received in Application No. _____ .

3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

* See the attached detailed Office action for a list of the certified copies not received.

14)☐ Acknowledgment is made of a claim for domestic priority under 35 U.S.C. § 119(e) (to a provisional application).

a) ☐ The translation of the foreign language provisional application has been received.

15)☐ Acknowledgment is made of a claim for domestic priority under 35 U.S.C. §§ 120 and/or 121.

Attachment(s)

1) ☒ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☒ Information Disclosure Statement(s) (PTO-1449) Paper No(s) 12 .
4) ☐ Interview Summary (PTO-413) Paper No(s). _____ .
5) ☐ Notice of Informal Patent Application (PTO-152)
6) ☐ Other: _____

DETAILED ACTION

Applicant's arguments with respect to the rejection of claims 36-38 under 35 USC § 112 have been fully considered but they are not persuasive. Additionally the formula used by applicant implies a chemical compound and A@Cn is the proper agreed upon way to express encapsulated material. Furthermore, the use of the formula is unnecessary as the limitation that the material A is encapsulated is already stated. The rejection is maintained in its entirety and incorporated herein by reference.

Applicant's arguments with respect to the rejections under 35 USC § 103 have been fully considered and are persuasive. It is noted by the examiner that the limitation of closing the openings by forming "passivation layers" is interpreted to exclude the closing of the openings by restoring the carbon cage. Accordingly the rejections are withdrawn. However, upon further consideration, a new ground(s) of rejection is made in view of the new interpretation.

Claim Rejections - 35 USC § 103

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

Claims 1-3, 10-11, 15, 20-21, 23, 26-28, 32, 39-41, 43-51, and 71 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan et al (USPN 5,457,343) in view of Hiura et al

(USPN 5,698,175) and further in view of Satishkumar et al. ("Novel experiments with carbon nanotubes…").

Ajayan discloses a process by which a nanometer sized carbon tubule enclosing a foreign material, except for carbon, may be produced. This process includes the steps of producing a nanotubule, opening a top portion of the tubule, introducing a foreign material into the center hollow through this opening, and closing the tubule. Ajayan discloses that the carbon tubules "may comprise a single cylinder of a graphite carbon monoatomic sheet" or alternatively "may comprise a plurality of cylinders of the graphite monoatomic sheets" (column 2, lines 45-59). Thus, the nanostructure may comprise SWNTs or MWNTs. Ajayan teaches that useful foreign materials to be introduced into the nanometer sized carbon tubule include alkali metals such as: lithium, sodium, potassium, rubidium, and cesium as well as magnetic metals such as iron, cobalt, and nickel (column 3, lines 21-35).

Regarding claims 27-28, Ajayan teaches that the foreign material may be introduced by evaporation on the top of the tubule or alternatively by contacting the top of the tubule with the gaseous compound including the foreign material (column 3, line 60 to column 4, line 3). Additionally Ajayan teaches that the nanotube may be opened by exposing it to a reactive liquid. Ajayan does not specifically teach the reactive liquids which may be applied. However, Satishkumar teaches that carbon nanotubes may be opened by exposure to a reactive liquid, including concentrated acid. Official Notice is taken that sonication is a well-known and often used method of mixing nanotubes and acids to provide for efficient exposure. It would have been obvious to one of ordinary skill at the time of invention to use the reactive acid liquids of Satishkumar in the process of Ajayan in order to open the carbon nanotubes.

Ajayan does not teach a step of purifying the tubules. However, Hiura et al. teach a similar process by which carbon nanotubes are purified, uncapped (opened), and filled. Regarding claims 10-11, the purification stage of Hiura involves acid reflux followed by filtration. This can be seen in Example 1, where the nanotubes are refluxed in a mixture of sulfuric acid and nitric acid and then filtered by a glass filter (column 4, lines 49-55). It is also seen in Example 5. The purification is performed in order to provide high-quality carbon nanotubes, which do not contain nanoparticles and amorphous carbon and which are required in order to achieve desired properties (column 2, line 64 to column 3, line10). It thus would have been obvious to one of ordinary skill at the time of invention to include the purifying step of Hiura in the process of Ajayan in order to provide high-quality products.

Ajayan teaches that the nanotubes may be closed by known processes. Satishkumar teaches that the filled nanotubes may be closed by "passivation layers" formed through the dispersion of the filled nanotubes in a solvent, such as methanol, ethylene glycol, and propylene glycol. It would have been obvious to one of ordinary skill at the time of invention to use the closing process of Satishkumar in the process of Ajayan because it is a known method for the closing of filled carbon nanotubes.

Regarding claims 39-44 and 47-48, the limitations are viewed as product by process limitations. Therefore, because no difference is seen between the processes of Ajayan in view of Hiura and taken with Satishkumar and that of the applicant, it is expected that the products will have equivalent properties.

Regarding claims 45-46, the use of a specific range concerning the amount of intercalant deposited in the carbon nanotubes is seen to be the optimization of a known process, which could

have been determined through routine experimentation and which is held to be obvious by *In re Boesch* 205 USPQ 215.

Claims 4, 17-19, 35, 52, and 63-64 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claims 1 and 49 above, and further in view of Jin et el (USPN 6,283,812).

Ajayan does not provide for the directional growing of the nanotubes on a support. However, Jin, in order to provide an array of nanotubes which contains a high directional density and thus efficient field emission properties, teaches a means of growing an aligned carbon nanotube ensemble by methods including CVD, arc discharge, and laser ablation. It thus would have been obvious to one of ordinary skill at the time of invention to grow nanotubes under a directional control in the process of Ajayan in order to ensure a high directional density and efficient field emission properties.

Regarding claim 17, Satishkumar teach a method whereas the tubes are sonicated in an acid medium, thus cutting the tubes and leaving open ends. Jin teaches a method whereby a high energy beam, such as an ion beam, is used to truncate an ensemble of nanotubes, thus leaving open-ended tubes (column 5, lines 21-28). It therefore would have been obvious to one of ordinary sill at the time of invention to combine the process of Jin and the process of Satishkumar in order to provide opening of a higher percentage of the tubes.

Regarding claims 18-19, Ajayan teaches the use of etching the nanotubes under a reactive gas as a method of opening the tubes. Jin teaches an opening of the nanotubes whereby the tubes are etched with oxygen plasma (column 5, lines 24-28). It thus would have been obvious to one

of ordinary still at the time of invention to implement the use of oxygen plasma as the media with which the nanotubes are etched.

Regarding claims 35, 52, and 63-64, Ajayan teaches that the nanotube is available as its structural completeness for a device possessing a high mobility of electrons (column 4, lines 31-35). Jin teaches that this device may be put to use as the cathode of various articles, including microwave amplifiers and flat panel field emission displays. Cathodes containing nanotube emitters exhibit all of the properties advantageous to electron emission (column 9, lines 38-57). It is further noted that any nanotube product with the enhanced properties would be useful in any field-emitting device which is well known in the art. Thus it would have been obvious to one of ordinary skill at the time of invention to apply the nanotube products of Ajayan for use in the devices disclosed by Jin.

Claims 5 is rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Tanaka et al (USPN 5,951,832).

Ajayan's process provides for the growing, opening, filling, and closing of carbon nanotubules, it does not provide for the same process to be applied to giant fullerenes, such as onions. Tanaka, however, teaches a process whereby an ultrafine particle is enclosed by a fullerene using an electron beam to drive the particle into the hollow center. Because this process has the same steps as the process of Ajayan, it would have been obvious to one of ordinary skill to apply the process of Ajayan on the onion fullerenes of Tanaka et al.

Claim 6 is rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Zettl et al (USPN 6,057,637).

Ajayan's process, as stated, is only applied to carbon nanotubules. It does not take into account the derivatives of the pure carbon tubes. Zettl, however, in order to take advantage of their electrical properties such as field-emission characteristics, uses a carbon nanotube derivative, specifically BxCyNz nanotubes. These tubes contain varying amounts of boron and nitrogen on their sidewalls, yet are capped in the same manner as pure carbon nanotubes. Because the process of Ajayan deals only with this capped portion, it would have been obvious to one of ordinary skill at the time of invention to perform the process of Ajayan on the nanotube derivatives of Zettl in order to take advantage of their field-emission characteristics.

Claims 7-8 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Homyonfer et al. (USPN 6,217,843).

Ajayan, as stated, is not applied to carbon nanotube derivatives. Homyonfer, however, teaches the intercalation of inorganic fullerene-like structures, such as nested fullerenes and nanotubes, with various metal atoms. These inorganic fullerene-like structures may take the form of MX_2 where M = Mo, W and S = S, Se. (column 1, lines 35-41 and column 2, lines 4-5). It thus would have been obvious to one of ordinary skill at the time of invention to apply the process of Ajayan to these inorganic nanotube structures in order to produce intercalated inorganic fullerene-like structures.

Claims 9 and 16 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Gao et al. ("Electrochemical intercalation of single-walled carbon nanotubes with lithium").

Hiura teaches a purification stage which provides that the amorphous carbon and nanoparticles are removed from the carbon nanotubes, however, he applies a process of acid reflux and filtration. Gao et al, in order to achieve the same result, purify the nanotubes by filtering the impurity phases through a membrane while keeping the nanotubes in suspension using a high-power ultrasonic horn. It thus would have been obvious to one of ordinary skill at the time of invention to substitute the purification process of Gao et al. in place of that of Hiura.

Regarding claim 16, the method of opening the nanotubes, as disclosed by Ajayan, is that of etching with a gaseous reactant. Gao et al, however, teach that mechanical ball-milling increases defect density and reduces the length of SWNTs by fracturing the graphite layers. They disclose that the ball-milling may facilitate ion diffusion into the nanotubes. It is known in the art that cutting carbon nanotubes does, indeed, lead to tubes with open ends. It thus would have been obvious to one of ordinary skill at the time of invention to replace the etching process of Ajayan with the ball-milling process of Gao et al. in order to cut the tubes, leaving open ends.

Claims 12-13 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Ebbesen et al. (USPN 5,641,466).

Whereby Hiura teaches the purification by an acid oxidizing agent followed by filtration for the purification of nanotubes, Ebbesen teaches that hydrogen peroxide is a useful purification agent. Therefore, it would have been obvious to one of ordinary skill at the time of invention to use the hydrogen peroxide of Ebbesen in the purification process of Hiura. Furthermore, the use of specific reaction temperatures and concentrations of hydrogen peroxide is viewed to be the optimization of a known process, which could have been determined through routine experimentation and is held to be obvious by *In re Boesch* (205 USPQ 215).

Claim 14 is rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Ebbesen et al (USPN 5,641,466) and Resasco et al (USPN 6,413,487).

The purification step of Hiura comprises refluxing the nanotubes in acid followed by filtering; it does, however, make note of extinguishing amorphous carbon and nanoparticles by an oxidation reaction.

Ebbesen, teaches the purification of nanotubes by heating to a temperature of between 600 °C and 1000 °C in an oxidizing atmosphere (column 1, lines 41-50). Resasco, on the other hand, teaches the burning off of amorphous carbon under treatment with a heated combustion gas containing oxygen. Resasco's oxidative purification takes place under temperatures of about 300 °C (column 11, lines 32-55). Therefore, it is seen that oxidative purification can take place at temperatures as low as 300 °C and as high as 1000 °C. Thus, it would have been obvious to one of ordinary skill at the time of invention to replace the purification process of Hiura with one comprising heating the nanotubes, in an oxidizing environment, to a temperature within the

range of 300-600 °C. Additionally, the use of this specific range is seen to be the optimization of a known process, which could have been determined through routine experimentation and is held to be obvious by *In re Boesch* (205 USPQ 215).

Claims 34 is rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Liu et al. ("Fullerene Pipes").

Regarding claim 34, Ajayan does not teach a washing step after any of the stages which follow purification. Hiura, however, does teach the necessity of clean and pure tubes in order to achieve desired properties. Liu et al disclose a method of further washing the nanotubes, in order to ensure that the cut nanotube pieces were molecularly perfect and chemically clean. It would have been obvious to one of ordinary skill at the time of invention to apply a further washing step to the already opened and/or filled tubes in order to ensure their purity and cleanliness.

Claims 22, 24-25 and 36-38 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Green et al (USPN 6,090,363).

Ajayan discloses that it would be useful to fill the carbon nanotubes with various metals, superconductors, magnetic materials, organic molecules, gases, and alkali metals. Green teaches that it would be useful to include metals or alloys in elemental form, including Mg, Ca, Sr, and Ba (column 2, lines 6-20). It is further disclosed that the metals may be in combined form, that halogens such as F, Cl, and Br are useful materials, and additionally that the deposited material

may be converted to the desired physical or chemical form after the filling process (column 2, lines 51-54). Therefore, it would have been obvious to one of ordinary skill at the time of invention to use halogen mixtures or metal halides as the deposited material. It is furthermore seen that the opening and filling procedure of Green occur simultaneously, with the oxidizing liquid, preferably nitric acid, also containing the material for deposition. It is therefore expected that the nitric acid may be encapsulated in the carbon nanotube in addition to the preferred intercalant material.

Claims 29-30 are rejected under 35 U.S.C. 103(a) as being unpatentable over Ajayan and Hiura et al. and Satishkumar et al. as applied to claim 1 above, and further in view of Moskovits et al. (USPN 6,129,901).

According to Ajayan, the opened nanotubes are filled by evaporation of the foreign species or gas transport of the foreign species. Another method is disclosed by Moskovits, whereby the open-ended tubes are filled by a reaction containing the foreign deposits and the nanotube material. He specifically discloses the use of Ni (column 4, lines 5-15), yet also teaches that numerous other metals may be deposited into the nanotubes, once formed. It thus would have been obvious to one of ordinary skill at the time of invention to fill the tubes by reaction of chemicals containing the foreign species and the purified tubes.

Regarding claim 30, it is further taught by Moskovits that "it will also be understood by those skilled in the art that the nanotubes may be filled with the metals by electrochemical deposition" (column 4, lines 48-50). It thus would have been obvious to one of ordinary skill to replace the filling mechanism of Ajayan with the electrochemical process of Moskovits.

Claim 53-59, 61 and 62 are rejected under 35 U.S.C. 103(a) as being unpatentable over Jin et al. (USPN 6,283,812) in view of Ajayan and further in view of Satishkumar.

Jin teaches a process beginning with the production of an aligned nanotube ensemble on a support surface. Regarding claim 54, Jin discloses that the aligned carbon nanotubes may be grown by methods including chemical vapor deposition (column 4, lines 60-65). He discloses next that the aligned carbon nanotubes are substantially encapsulated in a solid matrix, or insulating layer (column 7, lines 15-35). Regarding claim 55, he discloses that polymer encapsulates are useful. Jin also teaches opening of the tubes by truncating them. Regarding claims 56-57, this opening may be performed by etching with ion or plasma beams (column 5, 22-27). Regarding claim 58, it is also seen that Jin discloses a process of opening the nanotubes whereby a portion of the encapsulating matrix is removed (Figure 4). He does not, however, provide a process for filling or closing these tubes, although he makes note of the process (column 5, lines 60-62).

Ajayan, however, discloses a process for filling and closing carbon nanotubes which were previously opened. Regarding claim 59, Ajayan teaches a method of filling that involves the evaporation of the foreign species into the open end of the nanotube (column 3, lines 60-67). It would be obvious to include the filling process of Ajayan in the process of Jin because of the increased electron emission properties of filled nanotubes. Ajayan teaches that the nanotubes may be closed by known processes. Satishkumar teaches that the filled nanotubes may be closed by "passivation layers" formed through the dispersion of the filled nanotubes in a solvent, such as methanol, ethylene glycol, and propylene glycol. It would have been obvious to one of

ordinary skill at the time of invention to use the closing process of Satishkumar in the process of Ajayan because it is a known method for the closing of filled carbon nanotubes.

Regarding claim 62, Official Notice is taken by the examiner that hydrogen plasma is known to be useful for the cleaning of electron emitting carbon structures. It would have been obvious to one of ordinary skill at the time of invention to clean, or "activate" the carbon nanotube electron emitters of Jin using hydrogen plasma.

Claim 60 is rejected under 35 U.S.C. 103(a) as being unpatentable over Jin, Ajayan, and Satishkumar as applied to claim 53 above, and further in view of Moskovits et al. (USPN 6,129,901).

While Ajayan discloses a means of filling the tubes by evaporation of the foreign species, Moskovits notes that open-ended nanotubes may be filled with metals by electrochemical deposition (column 4, lines 48-50). It therefore would have been obvious to one of ordinary skill at the time of invention to replace the evaporation procedure of Ajayan with the electrochemical process of Moskovits.

Allowable Subject Matter

Claim 33 is objected to as being dependent upon a rejected base claim, but would be allowable if rewritten in independent form including all of the limitations of the base claim and any intervening claims.

Conclusion

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Peter J Lish whose telephone number is 703-308-1772. The examiner can normally be reached on 9:00-6:00 Monday through Friday.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Stanley Silverman can be reached on 703-308-3837. The fax phone numbers for the organization where this application or proceeding is assigned are 703-872-9310 for regular communications and 703-305-9311 for After Final communications.

Any inquiry of a general nature or relating to the status of this application or proceeding should be directed to the receptionist whose telephone number is 703-308-0661.

PL
March 31, 2003

STUART L. HENDRICKSON
PRIMARY EXAMINER